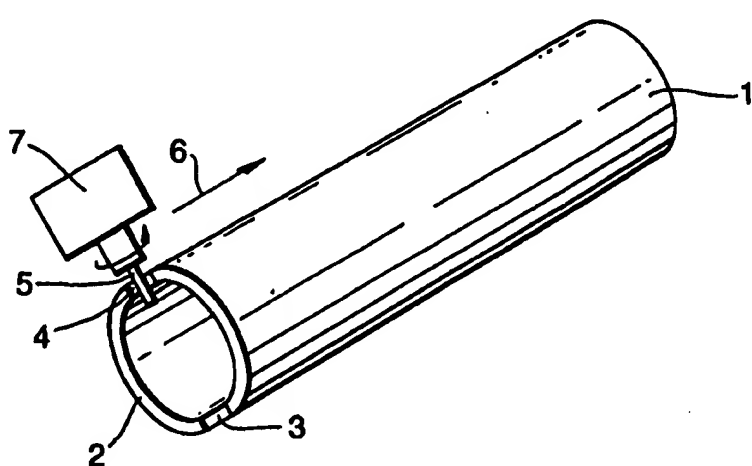




## INTERNATIONAL APPLICATION PUBLISHED UNDER THE PATENT COOPERATION TREATY (PCT)

<p>(51) International Patent Classification <sup>6</sup> : <b>B23K 20/12</b></p>	<p><b>A1</b></p>	<p>(11) International Publication Number: <b>WO 99/54081</b></p> <p>(43) International Publication Date: 28 October 1999 (28.10.99)</p>
<p>(21) International Application Number: PCT/GB99/01194</p> <p>(22) International Filing Date: 19 April 1999 (19.04.99)</p> <p>(30) Priority Data: 9808607.7                      22 April 1998 (22.04.98)                      GB</p> <p>(71) Applicants (for all designated States except US): THE WELDING INSTITUTE [GB/GB]; Abington Hall, Abington, Cambridge CB1 6AL (GB). SVENSK KÄRNBRÄNSLEHANTERING AB [SE/SE]; P.O. Box 5864, S-102 40 Stockholm (SE).</p> <p>(72) Inventors; and (75) Inventors/Applicants (for US only): HEDMAN, Tommy [SE/SE]; Ranvägen 5, S-187 70 Täby (SE). ANDERSSON, Claes-Göran [SE/SE]; Tjäderspelsv 25, S-155 30 Nykvarn (SE). FIELDING, Peter, Bernard [GB/GB]; Riverside Cottage, Hill Road, Great Sampford, Saffron Walden, Essex CB10 2RS (GB). NICHOLAS, Edward, David [GB/GB]; 106 Abbots Road, Haverhill, Suffolk CB9 0DH (GB).</p> <p>(74) Agent: GILL JENNINGS &amp; EVERY; Broadgate House, 7 Eldon Street, London EC2M 7LH (GB).</p>		<p>(81) Designated States: AE, AL, AM, AT, AU, AZ, BA, BB, BG, BR, BY, CA, CH, CN, CU, CZ, DE, DK, EE, ES, FI, GB, GD, GE, GH, GM, HR, HU, ID, IL, IN, IS, JP, KE, KG, KP, KR, KZ, LC, LK, LR, LS, LT, LU, LV, MD, MG, MK, MN, MW, MX, NO, NZ, PL, PT, RO, RU, SD, SE, SG, SI, SK, SL, TJ, TM, TR, TT, UA, UG, US, UZ, VN, YU, ZA, ZW, ARIPO patent (GH, GM, KE, LS, MW, SD, SL, SZ, UG, ZW), Eurasian patent (AM, AZ, BY, KG, KZ, MD, RU, TJ, TM), European patent (AT, BE, CH, CY, DE, DK, ES, FI, FR, GB, GR, IE, IT, LU, MC, NL, PT, SE), OAPI patent (BF, BJ, CF, CG, CI, CM, GA, GN, GW, ML, MR, NE, SN, TD, TG).</p> <p><b>Published</b> With international search report.</p>
<p>(54) Title: CORROSION RESISTANT ENCLOSURE AND METHODS FOR ITS MANUFACTURE</p> <p>(57) Abstract</p> <p>A method of fabricating a corrosion resistant enclosure for a radioactive material container, the method comprising friction stir welding at least two sections (1, 2) of copper or a copper alloy material together.</p> 		

**FOR THE PURPOSES OF INFORMATION ONLY**

Codes used to identify States party to the PCT on the front pages of pamphlets publishing international applications under the PCT.

AL	Albania	ES	Spain	LS	Lesotho	SI	Slovenia
AM	Armenia	FI	Finland	LT	Lithuania	SK	Slovakia
AT	Austria	FR	France	LU	Luxembourg	SN	Senegal
AU	Australia	GA	Gabon	LV	Latvia	SZ	Swaziland
AZ	Azerbaijan	GB	United Kingdom	MC	Monaco	TD	Chad
BA	Bosnia and Herzegovina	GE	Georgia	MD	Republic of Moldova	TG	Togo
BB	Barbados	GH	Ghana	MG	Madagascar	TJ	Tajikistan
BE	Belgium	GN	Guinea	MK	The former Yugoslav Republic of Macedonia	TM	Turkmenistan
BF	Burkina Faso	GR	Greece			TR	Turkey
BG	Bulgaria	HU	Hungary	ML	Mali	TT	Trinidad and Tobago
BJ	Benin	IE	Ireland	MN	Mongolia	UA	Ukraine
BR	Brazil	IL	Israel	MR	Mauritania	UG	Uganda
BY	Belarus	IS	Iceland	MW	Malawi	US	United States of America
CA	Canada	IT	Italy	MX	Mexico	UZ	Uzbekistan
CF	Central African Republic	JP	Japan	NE	Niger	VN	Viet Nam
CG	Congo	KE	Kenya	NL	Netherlands	YU	Yugoslavia
CH	Switzerland	KG	Kyrgyzstan	NO	Norway	ZW	Zimbabwe
CI	Côte d'Ivoire	KP	Democratic People's Republic of Korea	NZ	New Zealand		
CM	Cameroon			PL	Poland		
CN	China	KR	Republic of Korea	PT	Portugal		
CU	Cuba	KZ	Kazakhstan	RO	Romania		
CZ	Czech Republic	LC	Saint Lucia	RU	Russian Federation		
DE	Germany	LI	Liechtenstein	SD	Sudan		
DK	Denmark	LK	Sri Lanka	SE	Sweden		
EE	Estonia	LR	Liberia	SG	Singapore		

CORROSION RESISTANT ENCLOSURE AND METHODS  
FOR ITS MANUFACTURE

5       The invention relates to corrosion resistant enclosures for radioactive material containers and methods for their fabrication.

10       With the increasing proliferation of nuclear power stations and the like, there is an increasing need to provide secure and safe methods for disposing of radioactive, nuclear waste material. Material that cannot be reprocessed must be stored often for a considerable length of time, thousands of years or more. Until recently, containers for storing such radioactive waste were constructed of steel. Steel containers are  
15       manufactured in a number of parts which are then arc welded together leaving an opening to enable the waste material to be loaded into the container following which a closure member is arc welded to the container. The problem with steel containers is that there is the risk that with time  
20       they will corrode and radioactive material will leak.

      The most important requirement, therefore, for a radioactive material container is that it should remain intact for a very long time in the environment prevailing in a typical storage facility such as a deep repository.  
25       It must not be penetrated by corrosion in the ground water present in the rock of such a repository. It has therefore been proposed to provide a corrosion resistant enclosure to define at least an outer wall of the container. Copper and copper alloys have been identified as suitable materials  
30       for such an enclosure (see, for example, Svensk Kärnbränslehantering AB, 1996 Annual Report, Section 6.2). In order to provide strong corrosion resistance, the walls of these enclosures need to be as thick as possible and typical thicknesses are in the range 30-60mm.

35       It is believed that such materials at these thicknesses will provide very long term corrosion resistance. However, the enclosures need to be

manufactured in sections which are joined together. The joining process is critical since the long term corrosion resistance will be significantly reduced if the join itself is more prone to corrosion attack. Unfortunately, copper and copper alloys of the required thickness are very difficult to weld, primarily due to their high melting point (1083°C). To deal with this, it is therefore proposed to use electron beam welding which can achieve a very high integrity weld having similar corrosion resistance to the enclosure material itself. An example of an electron beam system which has been developed by The Welding Institute and which is suitable for this application is described in PCT/GB98/02882.

The tensile and creep strength is affected by the grain size of the material and the smaller the grain size, the better. Electron beam welding already achieves good corrosion resistance as mentioned above but can generate relatively large grains having dimensions in the order of millimetres. It is therefore desirable to try to improve still further the performance of the welded material while still achieving strong welds between the relatively thick sections.

In accordance with one aspect of the present invention, a method of fabricating a corrosion resistant enclosure for a radioactive material container comprises friction stir welding at least two sections of copper or a copper alloy material together.

We have found that friction stir welding can be used to achieve high integrity, corrosion resistant welds as an alternative to electron beam welding.

Friction stir welding is a method in which a probe of material harder than the workpiece material is caused to enter the joint region and opposed portions of the workpieces on either side of the joint region while causing relative cyclic movement (for example rotational or reciprocal) between the probe and the workpieces whereby frictional heat is generated to cause the opposed portions

to take up a plasticised condition; optionally causing relative translational movement between the workpieces and the probe in the direction of the joint region; removing the probe; and allowing the plasticised portions to consolidate and join the workpieces together. Examples of friction stir welding are described in EP-B-0615480 and WO 95/26254.

Friction stir welding (FSW) has been developed as a method for welding thin, light alloys such as aluminium and until now it was not considered possible to use this technique for welding much thicker materials, particularly of copper and copper alloys since it was well known that these materials are difficult to weld due to their high melting point and high thermal conductivity. However, we have found surprisingly that FSW can be used to weld thick copper and copper alloy materials. Although temperatures only reach 700-900°C, FSW is a solid phase technique so a good weld results. Furthermore, the resultant weld has a smaller grain size than can be achieved using electron beam welding. This is because the FSW process breaks up grains as they form and also allows the grains to cool quickly so that they have little chance to grow significantly. As a result, grain sizes of the order of microns are obtained. Tests have also shown that the hardness of the resultant weld is substantially the same as that of the parent material as is its tensile strength. FSW also allows the fabrication of corrosion resistant enclosures which generally are bulky and therefore heavy and so difficult to orient for electron beam welding. A typical nuclear waste container may weigh up to 25 tonnes.

Friction stir welding can accommodate a variety of weld orientations relatively easily while allowing a considerable tolerance in joint preparation as compared with electron beam welding.

Friction stir welding may be used to weld some or all sections of the enclosure. For example, a typical enclosure has a cylindrical form and will be made from two

semi-cylindrical halves together with a base and a closure member at each end. The two semi-cylindrical halves may be welded by electron beam welding as may the base while the closure member may be friction stir welded. This allows  
5 for benefits of both welding processes to be obtained while allowing the enclosure to be oriented with the opening facing upwardly where the closure member can be more easily welded using friction stir welding than electron beam welding.

10 Conveniently, the welding step comprises maintaining the sections of the enclosure stationary and moving a friction stir welding tool along a joint line defined between the sections. In other cases, however, the tool could be stationary and the sections moved.

15 Since the enclosure will normally be oriented with an upwardly facing opening for the purposes of loading radioactive material, in one example the closure member and the wall of the enclosure surrounding the opening are tapered so that the closure member can be supported by the  
20 enclosure prior to welding the closure member to the wall. In a second example, at least part of the closure member has a lateral dimension greater than the corresponding dimension of the wall of the enclosure surrounding the opening so that the closure member can be supported by the  
25 enclosure prior to welding the closure member to the wall.

The corrosion resistant material is copper or a copper alloy, the preferred material being nominally pure copper.

The enclosure could be used in certain circumstances to define a radioactive material container itself but  
30 typically, particularly where the container is to be buried deep underground, the container will be fabricated by providing a structure within the enclosure for holding the radioactive material. This could be a steel structure or the like to react against pressures exerted at depth and  
35 typically will be adapted to support fuel rods which are the commonest type of radioactive material to be stored.

In a typical storage process, an enclosure according to the invention is loaded with radioactive material through an opening; and a closure member is welded across the opening so that the radioactive material is fully contained within the container. Typically, the opening will face upwardly during the loading and welding steps although this is not essential.

In general, the enclosure will have a cylindrical form although other tubular cross-sections such as square and the like are possible.

Some examples of methods according to the present invention will now be described with reference to the accompanying drawings, in which:-

Figure 1 illustrates the fabrication of a cylindrical wall of an enclosure;

Figure 2 illustrates an end wall member welded to the cylindrical wall;

Figure 3 illustrates a closure member and the enclosure of Figure 2 just prior to welding and after radioactive waste material has been loaded into the container;

Figure 4 is a view similar to Figure 3 but illustrating a different example of a radioactive waste container;

Figure 5 illustrates graphically the variation in hardness value across a weld;

Figure 6a is a photomacrograph of a transverse section through an example of a weld;

Figures 6b and 6c show portions of the photomacrograph of Figure 6a in greater detail;

Figure 6d is a sketch of the photomacrograph of Figure 6a identifying the various regions of the weld;

Figures 7a-7d show examples of friction stir welding tools from the side and the end, for use in the examples shown in Figures 1 to 4; and,

Figure 8 shows details of the particular tool used to produce the weld shown in Figure 6a, the diagram not being to scale and illustrating dimensions in mm.

Figure 1 illustrates schematically two semi-cylindrical copper or copper alloy components 1,2, each having a length of about 5m and a thickness in the range 30-60mm. The two components are laid horizontally one above the other so as to define a pair of joint lines 3,4. The components 1,2 are then welded together by inserting a rotating friction stir welding tool 5 into the respective joint lines 3,4 and translating the tool along the joint line as indicated by an arrow 6. The tool 5 is mounted to a drive motor 7 which is mounted in turn to a support gantry (not shown) which traverses the motor 7 and tool 5 in the direction 6.

Following the welding operation just described, a cylindrical wall results which forms the main body of a corrosion resistant enclosure. One end of the enclosure is then closed by a copper or copper alloy end wall member 8 (Figure 2) having a thickness of 30-60mm. The end wall member 8 is inserted horizontally into one end of the cylindrical wall and defines an annular joint line 9 with the cylindrical wall. The end wall member 8 is then welded to the container by inserting the friction stir welding tool 5 into the joint line 9 and translating it around the joint line.

Following securement of the end wall member 8, an open ended enclosure results which is oriented vertically as shown in Figure 3 following which a steel support structure 10 is loaded into the enclosure through an upper opening 11 defined by a wall 12. The steel support structure 10 includes an array of longitudinally extending bores 13 into which spent fuel rods 14 are loaded under secure conditions through the opening 11. Any remaining space may be filled with an inert gas and then the opening 11 is closed by a copper or copper alloy closure member 15 having a thickness of 30-60mm. In order to locate the closure member 15 in



place, the wall 12 is tapered inwardly as shown in Figure 3 and a corresponding outer surface 16 of the closure member 15 has a corresponding taper. Figure 3 illustrates the components just before the closure member 15 is placed on the wall 12. Following that placement, the rotating friction stir welding tool 5 is inserted into the joint line defined by the surfaces 12,16 and translated around that joint line so as to friction stir weld the closure member 15 to the rest of the cylindrical enclosure.

10 In the example described, the friction stir welding tool 5 has been translated along the joint line but an alternative arrangement is possible in which the tool 5 is maintained stationary (while rotating about its axis) and the joint line itself is moved. It should also be noted that although all the joints in this example have been welded using friction stir welding, some could instead be welded using electron beam welding as mentioned above.

Figure 4 illustrates a second example. In this example, the two semi-cylindrical halves 1,2 are again welded together as shown in Figure 1 but in this case a base member 20 has a lateral dimension extending beyond the inner dimension of the cylindrical enclosure thus defining a joint line 21 which can then be friction stir or electron beam welded. In addition, in this example, a closure member 21 is provided having a similar form to the base member 20 which can again be friction welded along a joint line 22. It will be appreciated that the different types of base and closure members in Figure 3 and Figure 4 can be interchanged and mixed as necessary.

30 To illustrate the success of this invention, various welds have been made between copper alloys and these welds have been tested and examined. Figure 5 illustrates the variation of hardness value across a friction stir weld between two pieces of commercially pure, high conductivity copper. The region of the weld is indicated in Figure 5 (about 25-75mm) and it can be seen that the hardness value

(HV) across the weld is substantially unchanged from that in the parent material on either side.

In a second example, the tensile strength of the welded material was tested. In this example, 10mm thick friction stir welded copper plates were examined and found to have a tensile strength in the order of  $224\text{N/mm}^2$  which compares favourably with the tensile strength of the original 10mm thick plate of  $280\text{N/mm}^2$ . Furthermore, this tensile strength was found to be very consistent along the length of the weld.

The friction stir weld in various test samples has also been examined at the microscopic level to review grain size. One example is shown in Figure 6a which shows a photomacrograph of a 25mm thick friction stir welded copper plate. The photomacrograph clearly shows four main regions of differing grain structure which are identified for clarity purposes in the sketch 6d. Region X is the region of the copper plate which has been unaffected by the weld procedure. Region 1 is a thermally effected zone in which heat generated by the weld has caused slight modification of the grain structure. Regions 2, 3 and 5 are thermomechanically effected regions in which the structure has been plastically deformed with some areas of recrystallization. Finally, region 4 is a region of dynamically recrystallised material.

Examples of region 1 and a recrystallised portion of region 2 are shown under greater magnification in Figures 6b and 6c respectively. Comparison of the Figures clearly shows how recrystallisation of the grain structure in region 2 results in an increased grain size. Welds obtained so far have exhibited grain sizes in the order of  $80\text{-}600\mu\text{m}$  which compares very favourably with the preferred grain sizes in the order of  $180\text{-}360\mu\text{m}$ . These values should be compared with those typically found using electron beam welding which can vary from the original plate value of  $180\text{-}360\mu\text{m}$  to 4mm.

A variety of tools can be used for the welding tool 5 and some examples are shown in Figures 7 and 8.

The tools shown in Figures 7 and 8 each comprise a shoulder 31 and a tool pin 32 with a left hand thread form (for standard clockwise tool rotation).

The tool shown in Figure 7a has a pin with an oval cross-section while the tool shown in Figure 7b is provided with opposed flats 34 so as to define a paddle cross-section. Figure 7c is a variant of that shown in Figure 7b in which the flats 34 are replaced by re-entrant features 35. The tool shown in Figure 7d is similar to that shown in Figure 7a except that the thread has a progressively changing pitch and angle.

The tool shown in Figure 8 has a pair of flats 33 machined onto its sides. However, the cross-section of the pin 32 can be circular, or oval or flattened whereby the probe displacement volume is less than its volume of rotation so as to enable the easier flow of plasticised material.

In operation, the integral pin 32 and shoulder 31 assembly is rotated and plunged into a joint between the workpieces until the shoulder 31 penetrates the workpiece surface. When the pin 32 is first plunged, adjacent metal around it is friction heated as well as a small region underneath the pin 32 tip. The depth of penetration is controlled by the length of the pin 32 below the shoulder 31 by the tool.

Once in contact with the top surface of the substrate, the shoulder applies additional frictional heat to the weld region. In addition, the contacting shoulder 31, which can be machined to a variety of profiles to provide improved coupling, helps to prevent highly plasticised material from being expelled from the weld region.

When the rotating tool 31,32 is fully plunged, the thermomechanically softened material adopts a shape corresponding to that of the overall tool geometry. The heat affected zone is wider at the top surface where it is

in contact with the shoulder 31 and tapers down as the pin 32 diameter increases.

5       The combined frictional heating from the pin 32 and the shoulder 31 creates a highly plasticised "third body" condition around the immersed probe and at the workpiece surface. The plasticised material provides for some hydrostatic effect as the rotary tool moves along the joint helping plasticised material to flow around the tool. The plasticised weld material then coalesces behind the tool as  
10   the tool moves away.

CLAIMS

1. A method of fabricating a corrosion resistant enclosure for a radioactive material container, the method  
5 comprising friction stir welding at least two sections of copper or a copper alloy material together.
2. A method according to claim 1, comprising friction stir welding all sections of the enclosure together.
3. A method according to claim 1 or claim 2, wherein the  
10 enclosure defines an opening through which radioactive material can be loaded in use, one of the sections comprising a closure member for closing the opening.
4. A method according to claim 3, wherein the closure member and the wall of the enclosure surrounding the  
15 opening are tapered so that the closure member can be supported by the enclosure prior to welding the closure member to the wall.
5. A method according to claim 3, wherein at least part of the closure member has a lateral dimension greater than  
20 the corresponding dimension of the wall of the enclosure surrounding the opening so that the closure member can be supported by the enclosure prior to welding the closure member to the wall.
6. A method according to any of the preceding claims,  
25 wherein the welding step comprises maintaining the sections of the enclosure stationary and moving a friction stir welding tool along a joint line defined between the sections.
7. A method of fabricating a container for receiving  
30 radioactive material, the method comprising fabricating an open enclosure according to at least claim 3; and providing a structure within the enclosure for holding the radioactive material.
8. A method according to claim 7, wherein the structure  
35 is adapted to support fuel rods.
9. A corrosion resistant enclosure for a radioactive material container, the enclosure having at least two

sections of copper or a copper alloy material which have been friction stir welded together.

5 10. A radioactive material container for receiving radioactive material, the container comprising a corrosion resistant enclosure according to claim 9; and a structure within the enclosure for holding the radioactive material, the enclosure being open to allow radioactive material to be loaded into the structure.

10 11. A method of storing radioactive material, the method comprising providing a container according to claim 10, or a container fabricated according to claim 7 or claim 8, or a container having an enclosure according to claim 9 or fabricated according to any of claims 1 to 6; loading radioactive material into the container through an opening;  
15 and welding a closure member across the opening so that the radioactive material is fully contained within the container.

12. A method according to claim 11, wherein the opening faces upwardly during the loading and welding steps.

20 13. A method according to claim 11 or claim 12, wherein the closure member is friction stir welded to the enclosure.

14. A method according to any of claims 11 to 13, wherein the radioactive material comprises radioactive waste.

1/5

Fig.1.

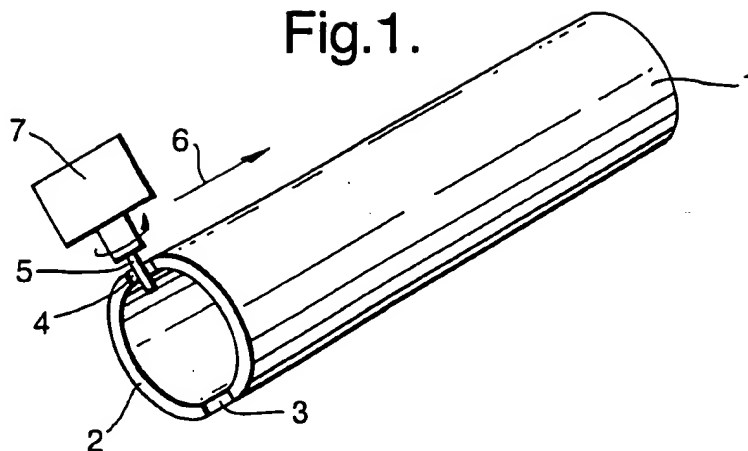


Fig.2.

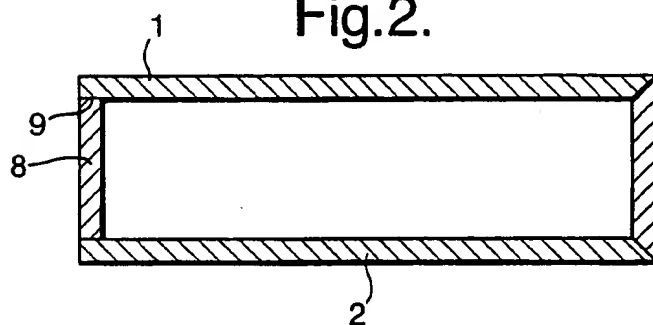


Fig.3.

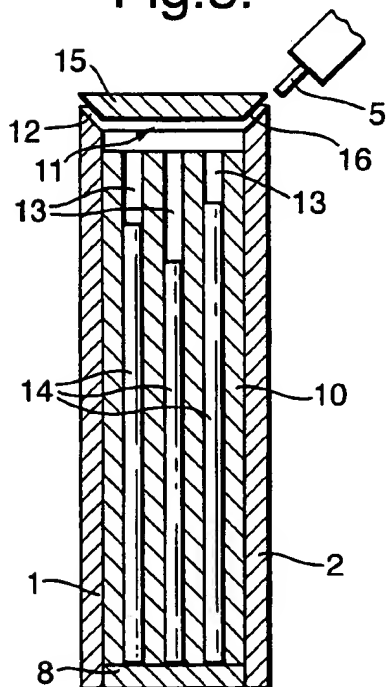
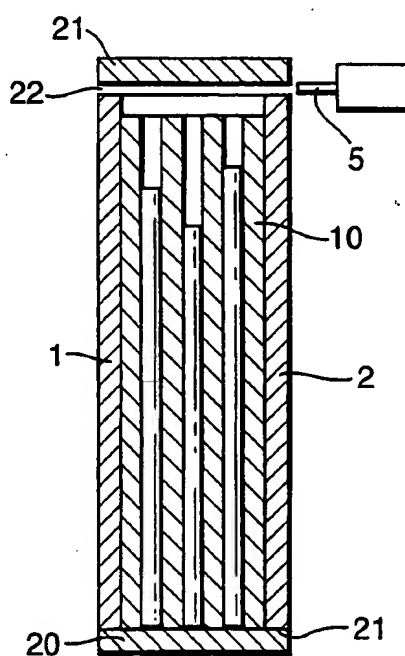
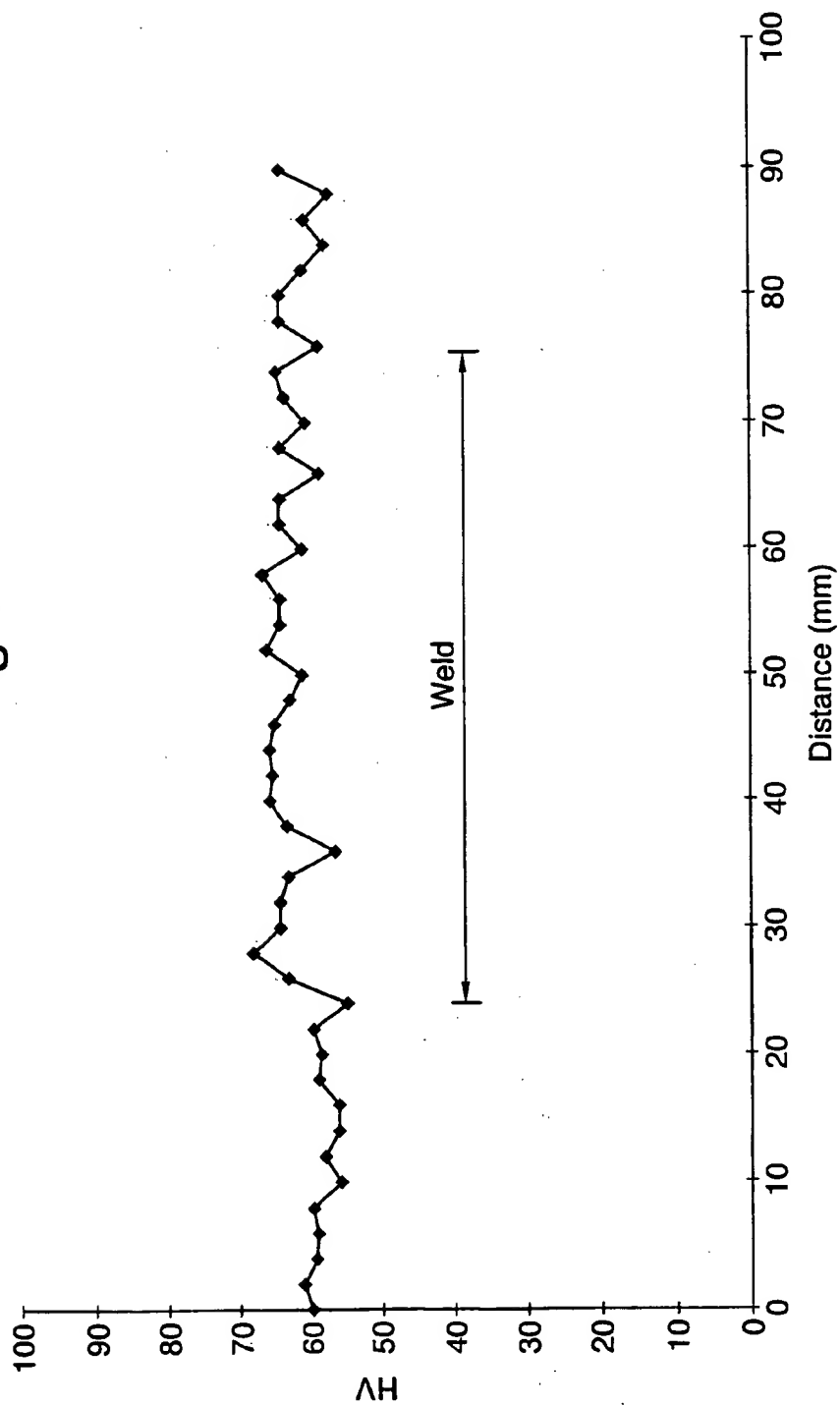


Fig.4.



2/5

Fig.5.





3/5

Fig.6a.

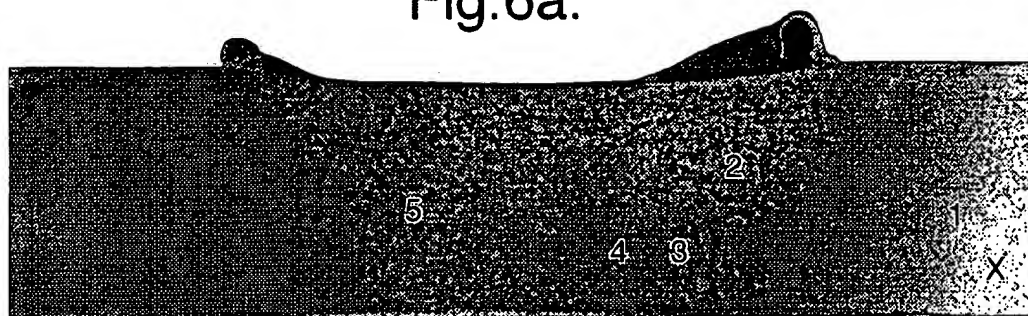


Fig.6b.

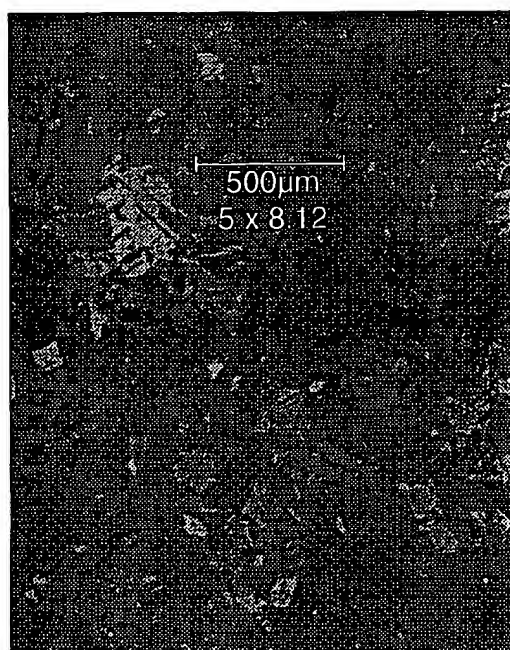


Fig.6c.

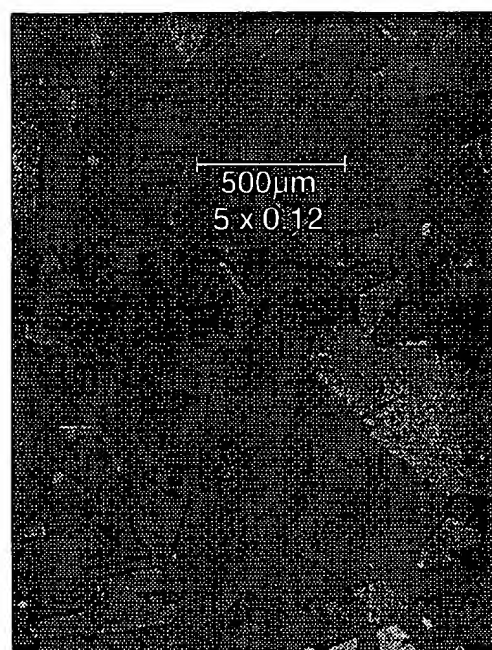


Fig.6d.

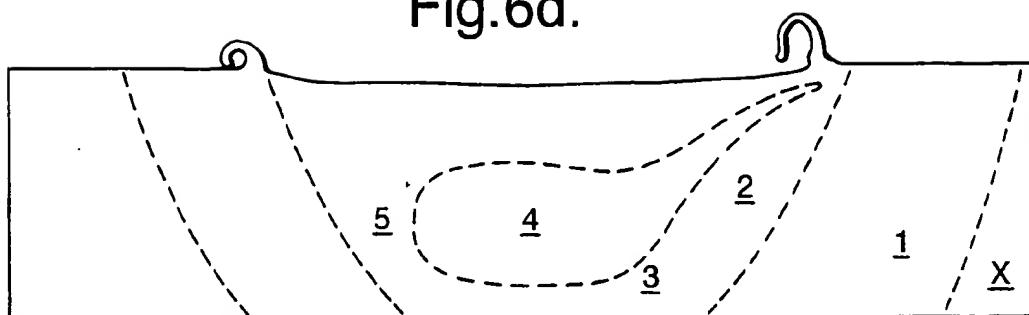


Fig.7a.

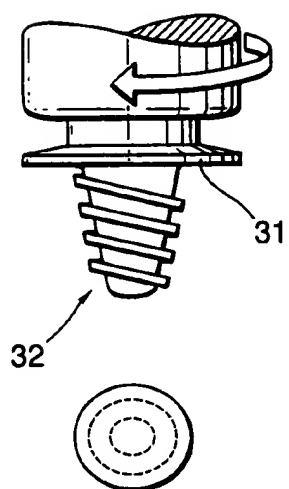


Fig.7b.

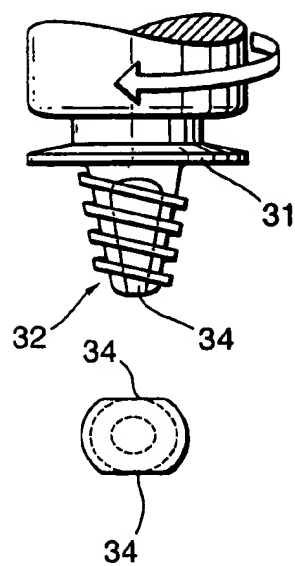


Fig.7c.

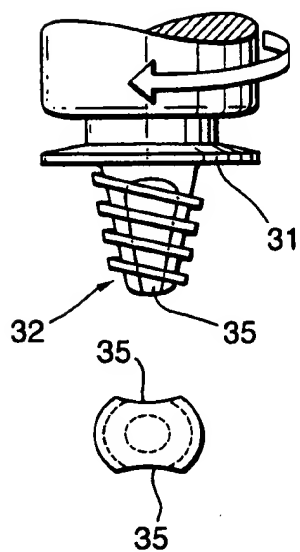


Fig.7d.

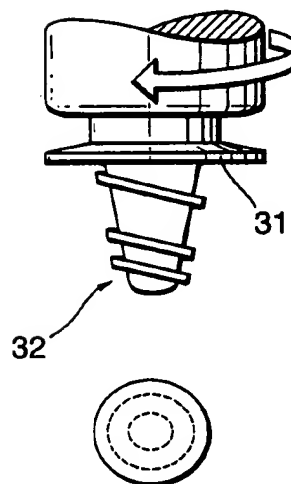
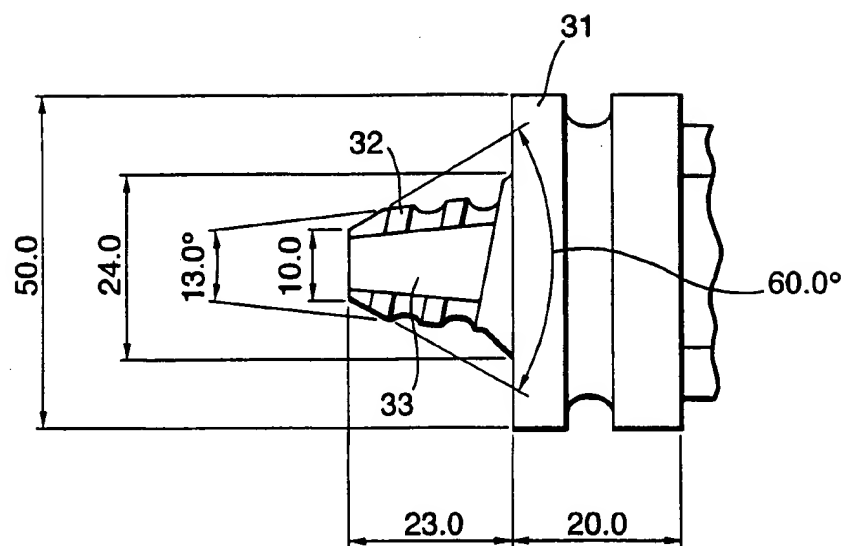


Fig.8.



# INTERNATIONAL SEARCH REPORT

International Application No

PCT/GB 99/01194

**A. CLASSIFICATION OF SUBJECT MATTER**  
IPC 6 B23K20/12

According to International Patent Classification (IPC) or to both national classification and IPC

**B. FIELDS SEARCHED**

Minimum documentation searched (classification system followed by classification symbols)

IPC 6 B23K

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)

**C. DOCUMENTS CONSIDERED TO BE RELEVANT**

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
Y	US 5 697 511 A (BAMPTON) 16 December 1997 (1997-12-16) column 2, line 50 - column 3, line 49; figures 1,2 ---	1-7,9
Y	US 4 542 846 A (MATSUI ET AL.) 24 September 1985 (1985-09-24) column 7, paragraph 1; figure 7 ---	5
Y	"Suitability of various materials for friction welding" METAL PROGRESS, vol. 119, no. 7, June 1981 (1981-06), page 49 XP002109790 ohio, USA table --- -/--	1-4,6,7, 9



Further documents are listed in the continuation of box C.



Patent family members are listed in annex.

\* Special categories of cited documents :

"A" document defining the general state of the art which is not considered to be of particular relevance

"E" earlier document but published on or after the international filing date

"L" document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)

"O" document referring to an oral disclosure, use, exhibition or other means

"P" document published prior to the international filing date but later than the priority date claimed

"T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention

"X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone

"Y" document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art.

"&" document member of the same patent family

Date of the actual completion of the international search

20 July 1999

Date of mailing of the international search report

04/08/1999

Name and mailing address of the ISA

European Patent Office, P.B. 5818 Patentlaan 2  
NL - 2280 HV Rijswijk  
Tel. (+31-70) 340-2040, Tx. 31 651 epo nl,  
Fax: (+31-70) 340-3016

Authorized officer \*

Herbreteau, D

# INTERNATIONAL SEARCH REPORT

International Application No

PCT/GB 99/01194

## C.(Continuation) DOCUMENTS CONSIDERED TO BE RELEVANT

Category	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	<p>EP 0 833 117 A (AUTOMOTIVE FLUID SYSTEMS INC.) 1 April 1998 (1998-04-01)  claim 8; figure 3  -----</p>	1-14

1